Collective lasing from a linear array of dielectric microspheres with gain

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Abstract: We experimentally study the optical emission behavior of a linear array of dielectric microspheres with gain. The microspheres are randomly arranged and well-separated, and can only couple via radiative modes. We observe resolution-limited, ultra-narrowband modes in the longitudinal emission, which constitutes collective lasing from the entire array, inferred from the observation of a lasing threshold. The lasing modes show wavelength selectivity, wherein the lasing probability is large only in specific frequency bands while being inhibited at other wavelengths, a behavior which is independent of the degree of configurational randomness. Analysis of the frequency bands indicates the participation of Fabry-Perot resonances of the individual microspheres in the collective emission.

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References and links
Collective optical phenomena are of a significant research interest currently [1]. They refer to phenomena that arise from the interactions between photonic components, with optical properties differing considerably from the individual components. An important motivation towards the study of collective phenomena is to obtain device functionality that is higher than that of the individual components [2]. Of late, the collective behavior of optical microcavities has seen a surge of interest [3–8]. For instance, collective behavior of linear chains of optical resonators, termed as ‘coupled resonator optical waveguides (CROWs)’, have been shown to offer waveguiding with interesting dispersion characteristics [9]. The existence of two collective modes, namely evanescent coupled and nanojet-induced coupled, have been shown in microsphere chains [10]. Efficient transport via the latter modes have been experimentally demonstrated [11]. The effect of size disorder on transport in linear chains has been addressed [12].

Apart from passive resonators, the collective emission of active resonators is also being studied. Theoretical studies have investigated linear or circular arrays of amplifying microcavities for low-threshold lasing [13, 14]. Experimentally, photonic molecule lasing has been demonstrated in bisphere configurations and semiconductor microdisks [7, 8]. However, the emission of multiple active microcavities has still not been addressed.

Furthermore, existing studies have focussed on microresonators that are either touching, or very closely spaced to enable strong coupling of the evanescent modes. Hitherto, the investigations have not addressed the scenario wherein the microcavities are separated from each other by several wavelengths, understandably due to the large radiative losses. Such radiative optical losses can possibly be overcome by the existence of gain. Nonetheless, to our knowledge, there have been no attempts to study linear chains of physically well-separated, amplifying microcavities, and to identify any resultant collective phenomena. In this work, we present experimental reports on the collective behavior of a linear array of amplifying spherical microcavities. The microspheres are separated by several tens of wavelengths, thereby admitting only radiative modes in the transport. We study both quasiperiodic and random configurations of such linear arrays. We find that, while the individual microspheres yield the expected Mie WGM lasing, collectively they manifest another lasing mechanism which gives ultra-narrowband modes. A statistical analysis over several spectra affirms a strong wavelength-dependence, in the sense that the modes are observed in selective intervals of wavelength. These observations point to the contribution of Fabry-Perot resonances of individual microspheres in the collective emission.

The experimental setup to generate the linear array and study the emission is shown in Fig. 1.
Monodisperse spherical microdroplets were created using a vibrating orifice droplet generator (VODG) [15]. The VODG has been traditionally used to study Mie scattering cross-sections and to make Mie lasers based on resonant whispering gallery modes (WGM) [16,17]. The technique is based on perturbation of unstable liquid jets. When a liquid is forced through a narrow orifice under pressure, it emits as a liquid jet that is mechanically unstable and prone to disintegration under perturbation. A periodic perturbation, with the appropriate amplitude and frequency, induces the jet to break up into equal-sized microdroplets. In our case, the liquid was a solution of Rhodamine 6G in Methanol at a concentration of 1 mM. The perturbation was applied by a piezo-electric gate, driven by a periodic signal, at the end of a microcapillary. We obtained microdroplet diameters within the neighborhood of 18 µm. The microdroplets descended under gravity in air in a linear configuration. Depending on the pressure and perturbation frequency, the configuration ranged from nearly periodic (referred henceforth as ‘quasiperiodic’) to random. Exact periodicity with a spacing within $\lambda/2$ could not be claimed because of the inherent dynamic nature of the system. The total length of the array emitted from the microcapillary was about 2-3 mm. In the central region of about 1 mm, the microdroplets were spherical and monodisperse. This length corresponded to about 25-40 microdroplets, depending upon the diameters and inter-droplet separation. The array was illuminated from the transverse direction with a pulsed Nd:YAG laser ($\lambda=532$ nm, pulsewidth $\sim 25$ ps). The resulting emission was spectrally analyzed by a 50 cm focal length spectrometer. The mirror M2, depicted in Fig. 1, collected the emission at an angle of $\sim 5^\circ$ from the longitudinal (along the array axis) direction, hereafter referred to as longitudinal emission. As shown in the inset, Lens L1 could be positioned to enable simultaneous collection of transverse and longitudinal emission. L1 collects the transverse emission directly from the array, while the longitudinal emission is collected from the image of the array in mirror M2. Lens L2 imaged the array on a CCD. Since the laser irradiated the array at a rate of 10 Hz, the array configuration was different at each excitation pulse.

Figure 2 illustrates the characterization of the array. Image (A) shows one quasiperiodic configuration of microdroplets. Plot (B) shows a typical WGM spectrum from an array, as measured from the transverse emission. The Q-factor of the WGM mode of a single mi-
microdroplet could not be resolved, and the width of the peaks originates from a slight inevitable polydispersity in the array. The microdroplets were sized using the following procedure. First, an estimate for the diameter $a$ was obtained from $a = (\lambda^2/\pi\delta\lambda)(\tan^{-1}\sqrt{m^2-1}/\sqrt{m^2-1})$, where $\delta\lambda$ is the free spectral spacing in the WG modes and $m$ is the refractive index of the droplet medium [18]. Next, a best fit was performed in the vicinity of this estimate by using the Mie theory, as shown by the red curve in plot (B), to obtain the accurate diameter. By tweaking the frequency of the piezo-electric gate, the array could be driven into a random configuration (image (C)). The configurational randomness is quantified by the histogram of (center-to-center) droplet separations, which shows a nearly Gaussian distribution with a spread of $> 20 \mu m$.

The emission properties of the system are summarized in Fig. 3. Fig 3(A) illustrates three
spectra obtained from a quasiperiodic configuration at three different excitation pulses with the same energy, $E_p = 0.55$ µJ. These spectra were measured from the longitudinal direction, assisted by the mirror M2 shown in Fig 1. The observed spectra entirely arose from collective emission, and no, or very feebly, signature of WGM modes was seen in the longitudinal direction. The spectra are generally multimode, with each spectrum showing narrowband lasing modes with a resolution-limited bandwidth of $\sim 0.2$ nm, implying a quality factor of at least $\sim 3000$. The number of modes was observed to increase with increasing excitation energy. Figure 3(B) illustrates the same from the random configuration at the same pump energy. The general characteristics of the spectra remained the same, despite the large deviation from periodicity. Figure 3(C) shows two spatio-spectral datasets (a) and (b), as captured by the spectroscopic CCD (CCD1) for two different excitation pulses. Top panels in the two sets display the spectral peaks discussed in Fig 3(A), while the middle panels show the corresponding bright spots as grabbed on the spectroscopic CCD. The bottom panels show the simultaneous transverse emission. It can be clearly seen that, at the positions of the spots in the middle panel, bright vertical streaks are observed. These vertical streaks extend over the entire illuminated array of microspheres. This light comprises the scattered fraction of the longitudinal mode. The transverse emission also prominently exhibits the WGM resonant modes (marked by the yellow arrows). The WGM modes do not change from pulse to pulse as they arise from individual microdroplets, whose diameters remain invariant. The collective modes (the vertical streaks), on the other hand, fluctuate from pulse to pulse, as they are sensitive to the instantaneous configuration. Figure 3(D) shows the variation of output intensity as a function of the excitation intensity, in the quasiperiodic system. The output intensity was an average over all modes in 100 spectra. The black curve shows the behavior of the collective emission, and a clear lasing threshold can be identified at $E_p = 0.12$ µJ, above which the output intensity diverges linearly. The curve for WGM lasing (red curve) had a lower threshold, and could not be captured in this experiment. Threshold behavior, indicative of the onset of lasing, was also observed in the random configuration. Although the experimental setup could not resolve the WGM mode of the individual microdroplet or the collective mode, the fact that WGM lasing is initiated first indicates a higher Q-factor for the WGM modes. Besides, the gain volume of the individual WGM mode is smaller than the collective mode, so inversion is reached rapidly in the former case.

The wavelength sensitivity in the phenomenon is not evident in the individual spectra, since they fluctuate from pulse to pulse. Nonetheless, a statistical study reveals the same, as seen in Fig. 4. Figure 4(A) shows the histogram of the lasing wavelengths from the quasiperiodic array, obtained over 100 spectra. Clearly, the histogram shows distinct bunches, implying that the ultra-narrowband lasing peaks occur only within specific intervals of wavelength, with other wavelengths having a very low probability for lasing at the said excitation energies. Fig 4(B) shows the same for the random array. Barring the fact that the histogram bunches moderately broaden out, the wavelength sensitivity is still clear in the histogram. Evidently, the wavelength sensitivity does not depend on the randomness of the array, but rather is a consequence of single microsphere resonance. Figure 4(C) traces the origin of this wavelength sensitivity. The plot reveals the contribution of Fabry-Perot resonances of single microspheres in the collective lasing. On the X-axis, we show the WG mode separation, which is the measurable quantity in our experiments. On the right Y-axis, the diameter calculated from the WGM separation is plotted as blue circles. For this diameter, we calculate the free spectral range of a FP cavity, $(\delta \lambda = \lambda^2 / 2md)$, formed between two opposite spherical surfaces of the same microdroplet. This FSR is shown as red circles, indicated on the left Y-axis. The black squares indicate the separation in the histogram bunches as measured in the experiments. The vertical bars on the square markers indicate the variation in FSR when multibunched histograms were obtained. As
easy transport via microscopic channels [21].

Another interesting scenario of collective microcavity lasing with leaky microcavities has been discussed in random lasers [20]. Given the configurational randomness of our system, it could be further investigated within the premises of random lasing. Finally, the fluidic nature of our system could be of great advantage in optofluidic systems, wherein liquids are preferred for easy transport via microscopic channels [21].

![Fig. 4](image)

Fig. 4. [A] Histogram of the lasing wavelengths from the quasiperiodic configuration. δλ indicates the separation between the bunches. [B] Same for the random configuration. [C] X-axis shows the WGM mode separation. The curve marked by blue circles depicts the calculated microdroplet diameter, as labeled on the right Y-axis. The left Y-axis shows the separation in the histogram bunches marked as dark squares. The red circles depict the calculated Fabry-Perot (FP) free spectral range for a FP resonator with the corresponding diameter. Inset: Schematic of the FP participation in the optical transport.

can be seen, there is an excellent agreement between the calculated FP free spectral spacing and the measured spacing over the range of diameters achievable in our system. Clearly, the system provides maximum amplification for those wavelengths that lie within the FP modes of the individual microspheres. This amplification is a result of the enhanced lifetime of these modes due to the FP resonances. Thus, only the modes whose frequency lies in the FP resonance profile are emitted by the linear array. This effect can be attributed to the geometrical configuration of the system, as follows. The initial fluorescence generated from a microdroplet is radiated in all directions. Only that fraction of the fluorescence traveling in the longitudinal direction experiences gain in the multiple microspheres, and is accordingly amplified as it propagates along the array. The longitudinally traveling light possesses wavevectors that can only excite the Fabry-Perot resonance of the microsphere, and hence undergoes wavelength selectivity according to the FP resonance profile of the microspheres. This effect is schematised in the inset of Fig. 4(C).

In summary, we have demonstrated a collective optical effect resulting from a random array of amplifying microspheres. This system exploits optical gain to overcome large radiative losses involved in coupling at a distance. We have shown ultra-narrowband lasing emission which originates from a co-operative behavior of the microspheres. Lasing modes with a high quality factor (> 3000) were observed. The frequency distribution of the lasing modes gives evidence of the contribution of Fabry-Perot resonances of the microspheres in the lasing.

Evidently, while an individual microsphere lases in a high-quality WGM, collectively they rely on Fabry-Perot resonances. The array offers higher functionality compared to the individual components in that the collective emission is directional as compared to isotropic emission from the individual microsphere, and the intensity is higher than the additive intensity of individual microlasers. Such a system could be evaluated for microphotonic device applications. It would be of interest to verify whether such systems can act as ACROWs (active CROWs) [19]. Another interesting scenario of collective microcavity lasing with leaky microcavities has been discussed in random lasers [20]. Given the configurational randomness of our system, it could be further investigated within the premises of random lasing. Finally, the fluidic nature of our system could be of great advantage in optofluidic systems, wherein liquids are preferred for easy transport via microscopic channels [21].